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Anthropogenic and natural methane fluxes in Switzerland synthesized within a spatially explicit inventory

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Abstract. We present the first high-resolution (500 m × 500 m) gridded methane (CH₄) emission inventory for Switzerland, which integrates 90 % of the national emission totals reported to the United Nations Framework Convention on Climate Change (UNFCCC) and recent CH₄ flux studies conducted by research groups across Switzerland. In addition to anthropogenic emissions, we also include natural and semi-natural CH₄ fluxes, i.e., emissions from lakes and reservoirs, wetlands, wild animals as well as uptake by forest soils. National CH₄ emissions were disaggregated using detailed geostatistical information on source locations and their spatial extent and process- or area-specific emission factors. In Switzerland, the highest CH₄ emissions in 2011 originated from the agricultural sector (150 Gg CH₄ yr⁻¹), mainly produced by ruminants and manure management, followed by emissions from waste management (15 Gg CH₄ yr⁻¹) mainly from landfills and the energy sector (12 Gg CH₄ yr⁻¹), which was dominated by emissions from natural gas distribution. Compared with the anthropogenic sources, emissions from natural and semi-natural sources were relatively small (6 Gg CH₄ yr⁻¹), making up only 3 % of the total emissions

in Switzerland. CH₄ fluxes from agricultural soils were estimated to be not significantly different from zero (between -1.5 and 0 Gg CH₄ yr⁻¹), while forest soils are a CH₄ sink (approx. -2.8 Gg CH₄ yr⁻¹), partially offsetting other natural emissions. Estimates of uncertainties are provided for the different sources, including an estimate of spatial disaggregation errors deduced from a comparison with a global (EDGAR v4.2) and an European (TNO/MACC) CH₄ inventory. This new spatially explicit emission inventory for Switzerland will provide valuable input for regional-scale atmospheric modeling and inverse source estimation.

1 Introduction

Most of the atmospheric methane (CH₄) produced in Switzerland results from anthropogenic activities. These emissions are well documented in the Swiss Greenhouse Gas Inventory (SGHGI, FOEN, 2013) that is updated and communicated to the UNFCCC on an annual basis. In contrast, the latest estimate for natural CH₄ fluxes including lakes,

wild animals, wetlands and forest soils (SAEFL, 1996) is outdated and was never compared with actual measurements taken in Switzerland. In 2011, the agricultural sector contributed 84.6 % to the total anthropogenic CH₄ emissions of 178 Gg CH₄ yr⁻¹, while the waste management and energy sectors added another 8.3 % and 6.8 %, respectively (FOEN, 2013). Since 1990, CH₄ emissions have decreased by about 20 % in Switzerland (FOEN, 2013). One reason is the decline in livestock numbers over the last 20 yr, mainly caused by changes in federal legislation. Emissions from natural gas distribution decreased due to the replacement of old infrastructure (Xinmin, 2004). However, this replacement process is now completed (Xinmin, 2004) and, combined with the projected higher demand for natural gas due to the new Swiss energy strategy (SFOE, 2012a), emissions are expected to increase again. Disposal of combustible waste in landfills has been prohibited since 2000 in Switzerland and therefore emissions from this source are decreasing (FOEN, 2013). Counteracting this trend, emissions from biogas production have more than doubled since 1990 and are projected to rise even further (FOEN, 2013). Thus, a reliable baseline inventory for CH₄ emissions from anthropogenic activities and natural processes is urgently needed.

Inventory estimates such as those presented in the SGHGI are based on numerous assumptions and statistical data that are associated with large uncertainties in several categories. Hence, validation by independent means is essential. Attempts have been made to constrain regional or national CH₄ emissions by atmospheric concentration measurements using boundary-layer budgets (Gallagher et al., 1994; Choularton et al., 1995; Fowler et al., 1996; Beswick et al., 1998; Wratt et al., 2001), inverse emission modeling (Vermeulen et al., 1999; Bergamaschi et al., 2005; Manning et al., 2011; Polson et al., 2011; Wennberg et al., 2012), or by discriminating individual sources with the help of the isotopic signature of CH₄ (Levin et al., 1999; Lowry et al., 2001; Miller, 2005). Studies of CH₄ fluxes in Switzerland mainly concentrated on measurements at a few selected sites and typically focused on improving process-level understanding rather than on providing representative numbers for national emission budgets. A first attempt was made by Hiller (2012, 2014b) to compare an earlier, preliminary version of the CH₄ inventory presented in this paper with CH₄ flux estimates based on aircraft measurements in a valley dominated by agriculture, the single most important CH₄ source in Switzerland. For this valley it was shown that the measured fluxes were in a similar range as the corresponding inventory values, but clearly more extensive evaluations using our inventory are needed.

In order to model the influence of CH₄ emissions on atmospheric concentrations, spatially explicit inventories are needed in addition to total national emissions (Bun et al., 2010). To disaggregate emissions to a higher spatial resolution, detailed knowledge of the location and the activity of each source is required, leading to additional uncertainty (Ciais et al., 2010). In recent years, the increasing targeting

of the atmospheric and inverse modeling community on the regional and urban scale has led to a clear trend towards high-resolution inventories. Currently, four different CH₄ inventories include Switzerland. EDGARv4.2, EDGAR-HTAP and TNO-MACC focus on anthropogenic emissions, NatAir considers only natural and biogenic emissions (details are shown in Table 1); thus, no inventory combines all CH₄ sources. Although these inventories have benefited from a considerable increase in resolution (e.g., EDGAR changed in 2009 from 1° × 1° to 0.1° × 0.1°), they are still limited to cell sizes of about 10 km × 10 km. For a spatially heterogeneous country such as Switzerland, this resolution is still too coarse to capture local variations.

The goal of this study was to produce the first gridded, high-resolution (500 m × 500 m) CH₄ inventory for Switzerland. Anthropogenic emission estimates followed the methodologies of the SGHGI (FOEN, 2013). National totals were either spatially disaggregated across Switzerland using the respective correlated geostatistical data with at least 500 m × 500 m spatial resolution, or generated in a bottom-up approach using emission factors (EFs). The SAEFL (1996) fluxes were updated and the total emissions for all relevant categories are reported together with their uncertainties. The additional uncertainty at the grid level introduced by the spatial disaggregation was estimated by comparing different inventories. For each source category, a brief review of recent research studies in Switzerland is presented and the results from field studies are compared with the inventory estimates where possible.

2 Data collection and processing

Our spatially explicit inventory is based on the SGHGI for anthropogenic emissions and additionally comprises natural fluxes. National emission totals were distributed onto a 500 m × 500 m grid according to correlated geostatistical data. Natural sources without up-to-date national totals available were up-scaled and spatially attributed using geostatistical data. Emission factors were adapted from the literature, including dedicated studies for Switzerland. The spatially explicit inventory was generated for the year 2011, which is the latest year available from the SGHGI. Sources are represented by positive numbers, sinks by negative numbers.

2.1 Swiss anthropogenic greenhouse gas inventory (SGHGI)

The latest submission of the SGHGI to the UNFCCC on 15 April 2013 reports greenhouse gas (GHG) emissions by sources and removals by sinks between 1990 (base year) and 2011 (FOEN, 2013). A detailed description of the institutional arrangements for inventory preparation, data sources and methodologies, uncertainty evaluations as well as quality assurance/quality control (QA/QC) activities are given in the

Table 1. Existing high-resolution CH₄ inventories that include Switzerland.

	EDGARv4.2	EDGAR-HTAP	TNO-MACC 2009	NatAir
Aim	Emission database for global atmospheric re-search	Emission database for global atmospheric re-search	Monitoring Atmospheric Composition and Climate (FP7)	Improving and Applying Methods for the Calculation of Natural and Biogenic Emissions and Assessment of Impacts on Air Quality (FP6)
Spatial resolution	0.1° × 0.1°	0.1° × 0.1°	1/8° × 1/16°	10km × 10km
Spatial coverage	Global	Global	Europe	Europe
Temporal coverage	1970–2008	2000–2005	2003–2007 and 2009	1997, 2000, 2001 and 2003 at hourly to annual time resolution
Included emissions	Anthropogenic emissions	Anthropogenic emissions	Anthropogenic emissions	Natural and biogenic emissions
Approach	Bottom-up inventory of internationally reported emissions	Official regional inventories like EMEP, gap filled with EDGAR v4.1	EMEP country totals checked for consistency, distributed according to geostatistical proxies	Bottom-up estimate
Reference	http://edgar.jrc.ec.europa.eu/index.php (last access: 1 Nov 2012)	Janssens-Maenhout et al. (2012)	Pouliot et al. (2012)	http://natair.ier.uni-stuttgart.de/ (last access: 1 Nov 2011), Friedrich (2007)

SGHGI (FOEN, 2013). The inventory preparation follows the reporting guidelines developed by the Intergovernmental Panel on Climate Change (IPCC) (IPCC, 1997, 2000, 2003, 2006). To estimate GHG emissions and removals following the IPCC methodology, three approaches differing in complexity (so-called tiers) can be used. The *Tier 1* methodology uses generalized default equations and parameters provided by the IPCC guidelines. *Tier 2* employs country-specific input data, providing more detail on the underlying processes with regional specificities. *Tier 3* is the most complex approach in terms of capturing dynamic processes and their spatial stratification, involving domestic measurements and/or modeling. The UNFCCC encourages parties to develop *Tier 3* methods for large sources and sinks as well as for those with temporal trends, the so-called key categories (IPCC, 2000). Switzerland is currently working to include more country-specific information for the next commitment period 2013–2020.

For the spatially explicit inventory, the eight strongest CH₄ sources out of a total of 620 listed in the SGHGI (FOEN, 2013) were selected, adding up to about 90 % of all anthropogenic CH₄ emissions. These eight sources include emissions from the agricultural sector (41 % from enteric fermentation of dairy cattle, 17 % from young cattle, 5 % from suckler cows, 2 % from sheep, 9 % from manure of dairy

cattle and 5 % of swine), the waste sector (5 % from landfills), and the energy sector (5 % from losses from natural gas distribution), where percentages in parentheses represent the share of the total 2011 anthropogenic emission estimate of 178 Gg CH₄ yr⁻¹. Additionally, we also compiled a spatial inventory of the emissions from wastewater treatment plants as these act as strong local sources.

2.1.1 Agricultural sector

The largest agricultural source in the SGHGI is *4.A Enteric Fermentation* followed by *4.B Manure Management* (the headings and numbers correspond to the official nomenclature for reporting, see Table 2). Agricultural residue burning is only a small source in Switzerland and reported in the sector *6. Waste*, whereas emissions from agricultural soils, rice production, and burning of savannas are negligible.

The 57 600 registered farms manage about one third of Switzerland's area (15 000 km² including Alpine pastures; FSO, 2013) and rear animals equivalent to 1 316 600 live-stock units (FSO, 2011).

Agricultural CH₄ emissions from livestock result from the microbial degradation of carbohydrates present in the rumen of ruminants, and to a lesser extent also in the hindgut of all herbivores (Jensen, 1996). Additionally, carbohydrates that

Table 2. Swiss CH₄ emissions in 2011, uncertainty estimate, and changes from 1990 to 2011 for the major source categories listed by the official Nomenclature for Reporting (NFR) codes. The provided uncertainty estimates follow the Tier 1 methodology (IPCC, 2000), represent half of the 95 % confidence interval expressed in percent (IPCC, 1997), and account for uncertainties in emission factors (EFs) and activity data for the individual level or category. Total national emissions exclude the Land Use, Land-Use Change and Forestry sector (LULUCF in italics) as well as International Bunkers (not shown), in accordance with the reporting requirements under the UNFCCC. Methods applied and EFs used are indicated (D = IPCC Default, T1 = IPCC Tier 1, T2 = IPCC Tier 2, T3 = IPCC Tier 3, CR = CORINAIR, CS = country-specific). All data for the anthropogenic sources are taken from the national Greenhouse Gas Inventory (FOEN, 2013), while CH₄ fluxes from the natural categories are based on estimates presented in this study. The categories indicated with an asterisk are included in our spatially explicit inventory.

CH ₄ source and sink categories	2011 [Gg GH ₄ yr ⁻¹]	Uncertainty [%]	Change since 1990 [%]	Methods	EFs
Anthropogenic	177.73	16	−20.2		
1. Energy	12.14	35	−58.9		
A. Fuel Combustion	3.89	35	−65.9	CS, T2, T3	CR, CS
3.b Transport; Road Transportation – Gasoline	1.00	35	−79.3		
4.b Other Sectors; Residential – Biomass	1.43	48	−68.7		
B. 2 Fugitive Emissions from Fuels; Oil and Natural Gas*	8.25	50	−54.4	T3, CS	CS
2. Industrial Processes (Chemical Industry)	0.41	30	−10.5	CS, T2	CS, D
3. Solvent and Other Product Use	NO				
4. Agriculture	150.43	18	−4.5		
A Enteric Fermentation*	119.48	18	−4.8	T2	CS
B Manure Management*	30.94	54	−3.2	T2	CS, D
5. Land Use, Land-Use Change and Forestry (Wildfires in Forest Land)	0.06	70	−84.9	T1	CS
6. Waste	14.72	48	−58.0		
A Solid Waste Disposal on Land*	8.61	58	−73.7	CS, D	CS, D
B Wastewater Handling*	0.48	30	115.6	D	CS, D
D Other	5.04	100	253.2	CS	CS
7. Other (Fire Damage in Buildings and Motor Vehicles)	0.03	30	3.9	T1	CS
Natural and semi-natural	5.7/−2.8	NA	NA		
Lakes and reservoirs*	2.3	NA	NA	See Sect. 2.2.1	
Wetlands*	2.3	NA	NA	See Sect. 2.2.2	
Wild animals*	1.1	NA	NA	See Sect. 2.2.3	
Agricultural soils	−1.5 to 0	NA	NA	See Sect. 2.2.4	
Forest soils*	−2.8	NA	NA	See Sect. 2.2.5	

are not digested and thus are excreted as volatile solids can subsequently be converted to CH₄ during manure management. Overall, the CH₄ production from enteric fermentation is primarily related to feed intake, standardized by using gross energy intake (GE) for inventory purposes. Intake differences quite reliably reflect variations in animal weight and performance (milk yield, growth, and pregnancy) and corresponding differences in CH₄ emissions (Soliva, 2006). However, variation in feed composition, i.e., in the substrates for the methanogenic archaea, is not accounted for. CH₄ production is assumed to decline when forage is partially replaced with concentrate in the ruminant diet (Beauchemin et al., 2008), but this reduction is often smaller than assumed, and about one third of this reduction may be subsequently compensated for by correspondingly higher manure-derived CH₄ emissions (Hindrichsen et al., 2006). Since Swiss ruminant diet types are mostly forage-based, CH₄ conversion rates measured in Switzerland are higher than IPCC (2006) default values (Zeitz et al., 2012). On the other hand, experiments on CH₄ emissions from Swiss manure manage-

ment result in lower emissions than currently estimated in the SGHGI using IPCC (2006) default values (Zeitz et al., 2012). In particular, emissions from liquid manure systems tend to be lower than those currently reported. The influence of animal genotype on the CH₄ emission potential is currently being discussed at the global level, but Swiss studies do not indicate significant differences between dairy breeds (Münger and Kreuzer, 2006). In conclusion, preliminary analyses suggest no significant change in CH₄ emissions from livestock by applying Swiss-specific EFs (Zeitz et al., 2012), as different under- and overestimates compensate each other. However, this conclusion does not yet consider the potential to reduce GHG emissions using different feeding measures (addition of lipids, plant secondary compounds, etc.; see, e.g., Beauchemin et al., 2008; Staerfl et al., 2012).

For our spatially explicit inventory, emissions were calculated from livestock numbers in 2007, aggregated by farm (agricultural establishment census 2007; FSO, 2009), and multiplied by animal-specific EFs from the Swiss national air pollution database (EMIS, Federal Office for the

Environment). Emissions for 2007 were then scaled to the 2011 value reported in the SGHGI. Following Swiss husbandry practice, most emissions were assumed to be produced in the stall (80 % for cattle, 20 % for sheep, 100 % for swine) and the remaining fraction in the pastures. The agricultural establishment census contains the location of the main farm building at one hectare resolution and was assumed identical to the stable and manure storage location (Kupper et al., 2010). Emissions in pastures were attributed to all grid cells covered by this land-use type (Swiss land-use statistics; FSO GEOSTAT, 2009) within the community of the respective farm. As part of Swiss farming practice, part of the livestock is moved to Alpine pastures in summer. Consequently, the CH₄ emissions produced there were also allocated in those Alpine pastures (≈ 4 % of the agricultural emissions included in the spatially explicit inventory).

2.1.2 Waste management

Within the waste management sector, CH₄ emissions originate mainly from 6.A *Solid Waste Disposal on Land*, 6.B *Wastewater Handling*, and 6.D *Other* processes, including composting, digestion of organic waste, and biogas upgrading.

Landfills

Gas production by decomposition of organic material in the anoxic waste body (typically 50–70 % CH₄ (v/v), 30–50 % CO₂, and trace amounts of other gases; Farquhar and Rovers, 1973) leads to advective and diffusive gas transport within the landfill pore system and eventually to emissions into the atmosphere (e.g., Franzidis et al., 2008).

Recent research activities related to landfill-derived CH₄ in Switzerland are limited to the Lindenstock municipal waste landfill near Liestal. This 12 ha landfill received $\approx 3.2 \times 10^6$ m³ of household, construction, and commercial waste between 1949 and 1994. Following closure, the waste was capped with a 2 to 2.5 m-thick cover soil consisting primarily of silty loam, and a gas-collection system was installed constructed of vertical and horizontal, partially screened, high-density polyethylene pipes. However, gas collection has not been attempted in recent years, and gas outlets remain closed with screw-cap lids. This is a unique feature of this landfill, as gas-collection systems on several other Swiss landfills are either in continuous operation or absent.

Experiments at Lindenstock compared CH₄ fluxes obtained by different methods (Gómez et al., 2009; Eugster and Plüss, 2010; Schroth et al., 2012) at or above the cover-soil surface, as well as below-ground fluxes. Results indicated that the studied section of the landfill was predominantly a net source of CH₄, with highest emissions close to the gas-collection outlets (daily mean fluxes ranging between 0.05 and 1.5 g CH₄ m⁻² d⁻¹) (Henneberger et al., 2012; Schroth et al., 2012). A net flux of up to -0.002 g CH₄ m⁻² d⁻¹ (up-

take) was usually observed away from the gas-collection outlets. CH₄ efflux from the waste body was highly variable over short distances and time. CH₄ oxidation activity in the cover soil was generally high, mitigating most of the produced CH₄, but also exhibiting substantial spatial variability (estimated to -1.92 to -64 g CH₄ m⁻² d⁻¹ in 2010), being strongest where efflux from the waste body was highest. Mitigation of landfill-derived CH₄ in the Lindenstock cover soil is mediated by a highly diverse, abundant methanotrophic community (Henneberger et al., 2012, 2013). Similar experiments during winter indicated stronger net CH₄ emissions (up to 2.5 g CH₄ m⁻² d⁻¹) from the studied landfill section (Ugolini et al., 2009). This was primarily attributed to a decrease in oxygen availability within the cover soil as a result of increased soil water content at shallow depths, but also to a decrease in temperature, which both adversely affected the CH₄ oxidation activity.

These results from Lindenstock are not sufficiently representative for estimating total landfill emissions in Switzerland, but they broadly agree with previous studies on landfills in Europe and the USA, reporting oxidation activities in a similar but higher range. Hence, cover soils in general provide an effective buffer for landfill-derived CH₄, mitigating emissions into the atmosphere as a result of the activity of methanotrophs (Whalen et al., 1990; Boeckx et al., 1996; Börjesson et al., 1998; Chanton et al., 2009; Gebert et al., 2009; Park et al., 2010).

Characteristics of the individual landfills, such as waste composition, dumping period, etc., were not available for Switzerland. Therefore, the national emissions of 8.6 Gg CH₄ yr⁻¹ as reported in the SGHGI FOEN (2013) were proportionally distributed to the hectares classified as landfills in the land use statistics (FSO GEOSTAT, 2009) for our spatially explicit inventory.

Wastewater treatment

CH₄ is produced in the sewage system as well as in the anoxic part of the wastewater treatment plant and the upgrading of sewage gas. To our knowledge, no direct wastewater CH₄ measurements exist for Switzerland; however, the CH₄ emissions can be estimated from the organic load in the wastewater. The chemical oxygen demand (COD) ranges from 100 to 110 g COD person⁻¹ d⁻¹, with one third each being aerobically respired, converted to CH₄, and remaining in the sewage sludge. A large part of the CH₄ produced is used for power and heat supply of the wastewater plant, and only about 10 % is directly emitted into the environment. The resulting EF of 0.9 g CH₄ person⁻¹ day⁻¹ lies within the range of reported conversion rates by Dealman et al. (2012) of 0.08 % to 1.2 % of kg CH₄ (kg COD)⁻¹. The amount of released CH₄ also depends on the sewage system (higher with long pipes at low inclination) and the plant type (higher with uncovered anoxic post-digester). Using an average EF of 0.9 g CH₄ person⁻¹ day⁻¹ and a 12

million population equivalent (Swiss population plus industrial wastewater load converted to additional population), annual emissions from wastewater collection and treatment result in about 4 Gg CH₄ yr⁻¹. However, a recent publication proposes a higher EF of 1.5 g CH₄ person⁻¹ day⁻¹, arguing that CH₄ production in the sewage system was underestimated (Wunderlin et al., 2013). The resulting CH₄ emissions would increase by 50 % to about 6 Gg CH₄ yr⁻¹. In contrast, the SGHGI is based on a completely different method reporting only 0.48 Gg CH₄ yr⁻¹, because the emissions are estimated from loss rates within the individual plant units and the total CH₄ used for energy or biogas production. Hence, emissions from tanks that are not connected to the gas system and emissions in the sewage are not included. To be consistent with the SGHGI, we proportionally distributed the 0.48 Gg CH₄ yr⁻¹ to the 854 plants in Switzerland (FOEN, 2012) based on their capacity expressed in population equivalents. However, emissions might turn out up to a factor twelve higher using alternative estimation approaches.

2.1.3 Energy sector

Total CH₄ emissions in the energy sector are divided into the subcategories *1.A Fuel Combustion*, where most emissions originate from road transportation and residential heating, and *1.B Fugitive Emissions from Fuels*. The latter emissions largely occur during the transmission of natural gas in pipelines (category *1.B.2 Oil and Natural Gas*; FOEN, 2013).

In Switzerland, 12.2 % of the total energy consumption was covered by natural gas in 2011 (SFOE, 2012b). Gas distribution in Switzerland includes ≈ 19 000 km of pipelines, from which 12 % are operated at pressures > 5 bar, 23 % between 1 and 5 bar, and 65 % < 1 bar. Another ≈ 6000 km of pipes guarantee the final distribution to the end user (SGWA, 2012). A large proportion of the transported gas transits Switzerland on the way from the production sites in northern Europe to Italy (Xinmin, 2004).

In the SGHGI, fugitive emissions of natural gas are estimated based on the amount of transported gas as well as on the infrastructure, namely the length, type and pressure of the gas pipelines (FOEN, 2013). Most emissions are assumed to occur during final distribution and consumption, while emissions from welded high-pressure pipes are assumed to be low (Xinmin, 2004). Therefore, emissions reported in the SGHGI were distributed close to the gas consumers for our spatially explicit inventory. Based on the national buildings and dwellings survey (FSO, 2010), the national emissions were proportionally distributed to those areas where natural gas is used for heating, i.e., to each 1 ha grid cell where at least two houses are heated with natural gas. The emissions were subsequently aggregated onto the 500 m × 500 m grid.

2.2 Natural and semi-natural CH₄ sources and sinks

The SGHGI only reports anthropogenic CH₄ emissions while natural and semi-natural fluxes are omitted, except for wildfires in the Land Use, Land-Use Change and Forestry (LULUCF) sector. CH₄ flux estimates reported in the SAFEL report (1996) were updated based on new EF and compiled in our spatially explicit inventory as described below.

2.2.1 Lakes and reservoirs

Approximately 3.5 % of Switzerland (1450 km²) is covered by lakes and reservoirs (FSO GEOSTAT, 2009), which can emit significant amounts of CH₄ (Bastviken et al., 2011). These CH₄ emissions can occur via four main pathways: (1) standard gas exchange at the air–water interface; (2) ebullition (bubbling) from aquatic sediments; (3) turnover of a stratified water column with storage of CH₄ in (anoxic) bottom water; and (4) transport by plants in the shallow littoral zones (Chanton and Whiting, 1995; Bastviken et al., 2004). Hydropower reservoirs have an additional fifth emission pathway as they release water for energy production. Often the turbine intakes of a hydropower dam are located in the CH₄-rich bottom water of a stratified reservoir, thus CH₄ can be emitted via degassing at the turbine or along the downstream river to which the water is released (Kemenes et al., 2007). The most important sink for CH₄ in aquatic environments is oxidation, which occurs mostly at oxic/anoxic boundaries in the sediment (e.g., Frenzel et al., 1990) or water column (e.g., Schubert et al., 2010) and can account for a significant reduction in total CH₄ produced by decomposition of organic material in a lake before the CH₄ reaches the atmosphere.

Measuring all of these CH₄ transport pathways and their spatiotemporal variability in a single lake requires immense effort. Thus, often only a subset of all possible pathways is directly measured, while others are either neglected or estimated from literature data. Truly accurate and validated models for estimating CH₄ emissions via all these pathways do not exist. An approach that can be used when attempting to estimate CH₄ emissions from a large amount of lakes without direct measurements is to use the equations proposed by Bastviken et al. (2004), which estimate diffusion, ebullition, and storage emissions based on comprehensive measurements from a collection of North American and European lakes (see Supplement for details). Bastviken et al. (2004) found significant relationships between the CH₄ emission estimates and measurable variables, such as lake area, dissolved organic carbon and phosphorus concentrations, water depth, and the volume of the anoxic fraction of the water column.

Following Bastviken et al. (2004), we estimated diffusion, ebullition and storage emissions of CH₄ from the lake areas of all major Swiss water bodies, but made the following modifications. (1) We tripled CH₄ emissions from lakes and

reservoirs shallower than 30 m based on direct measurements of emissions from a small, shallow lake in the low Alpine region, which indicated high rates of ebullition (Schubert et al., 2012). (2) For the hydroelectric reservoir Lake Wohlen on the Swiss Plateau, we directly used the emission estimate of DelSontro et al. (2010) that was based on a year-long measurement study and results in a value ten times higher than that obtained with the method of Bastviken et al. (2004). No adjustments to any other reservoirs were made as Lake Wohlen may not be a representative system within Switzerland. (3) We provided a rough temporal variability of ebullition emissions by assuming that ebullition occurs only during the warmest half of the year, as DelSontro et al. (2010) found a strong correlation between emissions and seasonal water temperatures. (4) Finally, we assumed ebullition not to be a relevant process in high Alpine lakes at altitudes above 1500 m a.s.l. since they receive only little organic input, have low water temperature, and quite low CH₄ concentrations in the water column (Diem et al., 2012). The resulting emission factors are summarized in Table S1. The above-presented modifications suggest that other factors in addition to those proposed by Bastviken et al. (2004) may need to be considered for estimating CH₄ emissions from lakes and reservoirs in the future.

The locations and areas of Swiss lakes and reservoirs were taken from the primary surfaces of the digital version of the Swiss topographical map at 1:25000 scale in vector format (VECTOR25; Swisstopo, 2004), while the depths for lakes > 0.1 km² were obtained from FOEN (2007a). We included all lakes that are contained in the Swiss water bodies information system (GEWISS; FOWG, 2000). Water depth data could not be found for 652 out of 798 lakes. Therefore, we assumed that the depth of lakes < 0.2 km² (678 lakes) is less than 30 m. Lake altitude was taken from the digital elevation model (FSO GEO-STAT, 2006). In total, we found that lakes > 0.1 km² emit 2.1 Gg CH₄ yr⁻¹, with a 21 % (0.4 Gg CH₄ yr⁻¹) share from hydroelectric reservoirs. Smaller water bodies contribute another 0.2 Gg CH₄ yr⁻¹.

2.2.2 Wetlands

Wetlands are the largest natural source of CH₄ globally, where it is produced by microbial decomposition of organic material under anoxic conditions. However, wetlands have become rare in Switzerland (0.5 %, 200 km², of the land area today compared to 6 % in 1800; FOEN, 2007b). In our study, we also considered wetland areas of a mixed ecosystem type and hence a ten times larger area (see Table S2). They are classified as wetlands on the basis of their high biodiversity, protected by the Swiss legislation on the protection of mires, rather than by their hydrogeological properties that would better reflect their characteristics in terms of CH₄ fluxes.

Most information on wetland CH₄ fluxes originate from the Arctic, boreal, and tropical zones, and it is not trivial to

translate those results to Swiss wetlands. An important complication is the fact that even in moist environments the vegetated surface may act as a net sink for atmospheric CH₄ when water saturation in the soil is limited to deeper layers or when drainage ditches lower the average water table (e.g., Moore and Roulet, 1993). In most cases it can be expected that periods where wetlands are a sink for CH₄ are restricted to a few warm and dry weeks a year, which reduces the overall annual CH₄ emissions from such ecosystems compared with permanently waterlogged wetlands.

In Swiss fens, CH₄ emission rates ranging from 100 to 330 mg CH₄ m⁻² d⁻¹ have been reported for the summer months (Alpine fen at Göschener Alp, Liebner et al., 2012). Constant emissions between 0.12 and 31 mg CH₄ m⁻² d⁻¹ were also found from glacier forefields with calcareous bedrock (Nauer et al., 2012), while mires on siliceous bedrock were either a weak source of CH₄ (38 % of all cases), neutral (31 %), or a CH₄ sink (31 %; -0.14 to -1.1 mg CH₄ m⁻² d⁻¹; Nauer et al., 2012). Even in the case of large emissions from calcareous glacier forefields, Nauer et al. (2012) observed that roughly 90 % of the CH₄ produced in the deeper soil was oxidized before it reached the soil surface and the atmosphere. This agrees with other studies, which indicate that in the top centimeters of the soil above the water table, where oxygen is abundant, most of the CH₄ produced by methanogenic archaea is oxidized and hence the flux of CH₄ to the atmosphere is substantially lower than what microorganisms produce (e.g., King et al., 1998).

For our spatially explicit inventory, CH₄ emissions from Swiss wetlands were estimated from the wetland areas in Switzerland and literature-based EFs available for different wetland types as summarized in Table S2. Different types of wetland areas were determined from the national inventories of raised bogs, fens and mires (FOEN, 2008b, 2010) as well as of riparian landscapes (FOEN, 2008a). Additionally, the wetland core and sprawl areas reported in the national ecological network (FOEN, 2011b) were included in the analysis since these contain additional wetlands of regional and local importance. For the emission estimate, polygons were mapped to a 100 m × 100 m raster. Grid cells classified by one of the different types of wetlands were subsequently multiplied by the corresponding EF. If a grid cell belonged to more than one wetland type, the one with highest priority was selected. The priority refers to the level of detail of the data set (e.g., specification of different zones within a wetland) and the importance of a wetland type for CH₄ emissions. As a final step, the data were averaged to the 500 m × 500 m grid. In total, Swiss wetlands are estimated to emit approximately 2.3 Gg CH₄ yr⁻¹.

2.2.3 Wild animals

Red and roe deer, Alpine chamois and Alpine ibex are the most abundant wild living ruminants in Switzerland. CH₄ emissions from these wild animals were estimated from the

animal population estimates at cantonal (state) level in 2011 (except for Canton of Jura: 2006 and Canton of Vaud: 2009) (FOEN, 2011a). We multiplied these animal populations by the respective species-dependent EF in SAEFL (1996) (see Table S3). The spatial distribution depends on the habitat of the animals. While red deer prefer dense and open forest, roe deer prefer dense forest. Alpine chamois prefer unproductive vegetation as well as rocks and scree, while Alpine ibex mainly thrive on rocks and scree. The respective land cover types were selected from the Swiss land-use statistic (FSO GEOSTAT, 2009). Additionally, the locations of these Alpine habitats were restricted to altitudes above 1500 m a.s.l. (FSO GEOSTAT, 2006).

The number of large wild animals (260 000 red and roe deer, Alpine chamois and Alpine ibex) in Switzerland (FOEN, 2011a) is substantially less than the 1 580 000 cattle (FSO, 2011) in the agricultural sector. Moreover, wild animals are smaller in size and show a smaller energy uptake than cattle. This results in a comparatively low emission estimate of 1.1 Gg CH₄ yr⁻¹.

SAFEL (1996) also reported substantial emissions of 2.8 Gg CH₄ yr⁻¹ from rodents. Radar measurements of the mice density on Swiss fields resulted in an average of 9000 mice km⁻² (AGFF, 2012), while the rabbit density was estimated as 2.7 rabbits km⁻² (Zellweger-Fischer, 2012). Scaled to the 10 500 km² agricultural area in Switzerland (FSO, 2011), this translates to ≈ 94.5 mio. mice and 28 350 rabbits. Multiplied by the EFs of 0.26 g CH₄ mouse⁻¹ yr⁻¹ (Jensen, 1996) and 80 g CH₄ rabbit⁻¹ yr⁻¹ (IPCC, 2006), the annual emissions result in 0.027 Gg CH₄ yr⁻¹, which is far less than previously assumed and does not represent a significant contribution to the emissions from wild animals. Hence, rodents were not included in our spatially explicit inventory.

2.2.4 Agricultural soils

Two counteracting processes – methanogenesis and methanotrophy – drive the net exchange of CH₄ between agricultural soils and the atmosphere. In Switzerland, the agricultural sector comprises typical crop production on arable land (18 %), comparatively large areas of grasslands (49 %) and Alpine summer pastures (33 %), adding up to 15 000 km², corresponding to more than one third of the total area of Switzerland (FSO GEOSTAT, 2009; FSO, 2011). Several studies conducted at managed grasslands and Alpine pastures in Switzerland have reported small CH₄ uptake rates by soils (Hartmann et al., 2010; Stiehl-Braun et al., 2011; Imer et al., 2013; Merbold et al., 2013), which is also supported by other studies (Mosier et al., 1991; Flessa et al., 1998; Ineson et al., 1998; van den Pol-van Dasselaar et al., 1999; Kammann et al., 2001). The CH₄ fluxes depend on multiple drivers such as water-filled pore space, soil and air temperatures, nutrient availability, management activity such as fertilizer application or tilling, and soil texture. These drivers are site-specific, but also change temporally at a sin-

gle site. Recent results from three grassland sites in Switzerland reveal large temporal and spatial variations in CH₄ fluxes from managed ecosystems (Imer et al., 2013), ranging from a small sink ($-1.37 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) to a slight source ($0.59 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) on a daily timescale, and averaging to an annual mean flux of $-0.21 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ and $-0.30 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ for two sites with almost year-round measurements (Imer et al., 2013). To the best of our knowledge, these are the only two year-round data sets that exist in Switzerland, leading to large uncertainties when up-scaling to the total area of managed agroecosystems. We expect the annual net CH₄ flux for Switzerland to range between 0 and $-1.5 \text{ Gg CH}_4 \text{ yr}^{-1}$ (EF: $-0.14 \pm 0.14 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$; Freibauer, 2003) for the 10 500 km² agricultural land, excluding Alpine pastures, being a small sink. This is comparable in magnitude with other natural fluxes, but does not significantly contribute to the total CH₄ budget of Switzerland. Since CH₄ uptake across the agricultural areas is highly spatially variable, we did not attempt to distribute this small CH₄ sink spatially across Switzerland in our study.

2.2.5 Forest soils

The net CH₄ flux of forest soils is again dominated by the two counteracting processes, methanogenesis and methanotrophy. The available literature suggests that forest soils generally are a larger CH₄ sink than agricultural soils due to higher soil gas diffusivity in these systems (Smith et al., 2000). CH₄ fluxes over Swiss forest soils have been investigated only very recently (Frey et al., 2011; Gundersen et al., 2012; Hiltbrunner et al., 2012). Interestingly, uptake rates of $-1.5 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ for forest soils were found, which changed to a CH₄ source of up to $2 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ when soils were compacted by forestry machinery (Frey et al., 2011). These soil emissions persisted for several years (S. Zimmermann, personal communication, 2012), but were limited to relatively small areas compared to the total forest extent. Effects of soil compaction were therefore not considered in our spatially explicit inventory.

To estimate the CH₄ uptake by Swiss forests, we followed a method developed by Hobi et al. (2011). Forest cover was derived from the land-use statistics (FSO GEOSTAT, 2009), and forest type information was taken from the 25 m × 25 m forest mixture data set (FSO GEOSTAT, 2004) and thereafter aggregated to 100 m × 100 m. CH₄ uptake rates differ significantly between evergreen ($-0.46 \pm 0.27 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) and deciduous forest soils ($-1.12 \pm 0.68 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$), according to the literature reviewed by Hobi et al. (2011). Forest areas were therefore multiplied by the uptake rate appropriate for the type of forest at a 1 ha resolution. For mixed forests an average rate was used. Finally, the data were averaged to a 500 m × 500 m grid. Overall, our estimate of CH₄ net flux of forest soils is $-2.8 \text{ Gg CH}_4 \text{ yr}^{-1}$ for 2011.

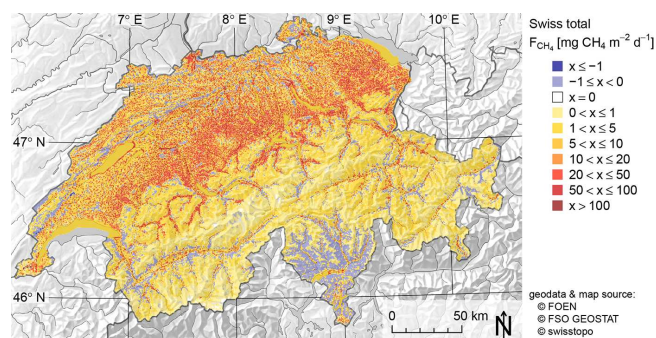


Fig. 1. Our spatially explicit Swiss CH₄ emission inventory including both anthropogenic and natural CH₄ sources.

3 Results and discussion

3.1 Spatially explicit CH₄ inventory

Anthropogenic emissions are strongly dominating total Swiss CH₄ emissions and mostly originate from agriculture (see Table 2). Hence, the highest emissions are observed in the southern part of the Swiss Plateau, an area dominated by livestock farming between the pre-Alps to the south and the Jura mountains to the north, covering approx. 30 % of Switzerland (Figs. 1 and 2a). Due to the proximity to the Alps, this region receives more precipitation than the rest of the Swiss Plateau and is therefore less suited for the production of vegetables and cereal, which are mainly cultivated in the northern and western parts of the Swiss Plateau. The central and northern parts of the Swiss Plateau are densely populated and consequently less land is dedicated to agriculture, which corresponds to relatively low emissions in this region. In the Alps, agricultural activity is concentrated on the valley floors. During the summer months, part of the livestock is moved to Alpine pastures for grazing to save the resources in the valley for the winter. This practice is part of the traditional Swiss three-stage farming system (Bätzing, 2003) and therefore CH₄ emissions can also be found in relatively remote areas of the Alps.

CH₄ emissions from waste management (Fig. 2b) are more abundant in regions with high population density. This also applies to the energy sector (Fig. 2c), where highest CH₄ emissions occur in urban areas because natural gas is distributed to private households for cooking and heating.

Natural and semi-natural CH₄ emissions from lakes, wetlands, and wild animals (Fig. 2d–f) as well as the uptake by forest soils (Fig. 2f) are considerably lower than anthropogenic emissions (Table 2). Natural lakes in Switzerland are remnants from previous glaciations. The largest lakes are located in the lowlands, while many small lakes are found throughout the country. Reservoirs are mainly situated in Alpine areas to exploit the descent for hydropower generation (Fig. 2f). Since the large wetlands in the floodplains were drained for agricultural use in the 19th and 20th centuries, the

highest emissions from this ecosystem type are limited today to shore areas and to hilly landscapes where agriculture is less favorable (Fig. 2d). Wild animals are more abundant in rural areas with continuous forests, the preferred habitat for many species. Alpine ibex and chamois also populate remote and sparsely vegetated mountainous areas (Fig. 2e). Forests cover mountain slopes up to the timberline, protecting from natural hazards. At lower elevations, forests were often converted into agricultural land during the last centuries, but some remained and are protected today by law. While wild animals living in the forest are a source of CH₄, the forest soil acts as a sink. Deciduous forests are limited to lower elevations, whereas evergreen forests dominate at higher elevations. Due to the lower uptake rate of evergreen forests, CH₄ uptake by forest soil tends to decrease with elevation.

Our spatially explicit inventory is available from <http://doi.pangaea.de/10.1594/PANGAEA.828262> (Hiller et al., 2014a).

3.2 Comparison with other inventories

The EDGAR v4.2 inventory and the TNO/MACC inventory in Fig. 3 show spatial distributions of total anthropogenic CH₄ emissions over Switzerland for the latest available years of 2008 and 2009, respectively. These maps can be qualitatively compared with the total emissions of our inventory presented in Fig. 1, where anthropogenic emissions make up more than 95 %.

The total emission of EDGAR v4.2 clipped to the domain of Switzerland amounts to 236 Gg CH₄ yr⁻¹ for 2008, consistent with the country total reported by EDGAR v4.2 for Switzerland. This total is almost 30 % higher than the 183 Gg CH₄ yr⁻¹ reported in the SGHGI for the same year. The TNO/MACC inventory adds up to 191 Gg CH₄ yr⁻¹ over the domain of Switzerland in 2009, which is close to the 180 Gg CH₄ yr⁻¹ in the SGHGI for 2009 (FOEN, 2013). The difference between EDGAR and TNO/MACC likely reflects the fact that EDGAR is an independent inventory applying its own methodologies for the collection of activity data, application of emission factors, and spatial allocation. The TNO/MACC inventory, in contrast, is scaled to total emissions reported by the individual countries. In both inventories, the spatial allocation of the emissions is based on different and less detailed geostatistical information than available in our study. Fig. 4 presents scatter plots of the pixel values of the EDGAR and TNO/MACC inventories versus our inventory mapped to the respective resolution of the coarser inventory. Scatter plots were created for both total emissions and for different categories separately. For the scatterplot, we used the subsectors that represent the emissions in our inventory best, while the comparison of Swiss totals per sector is based on all emissions of each sector reported in the respective inventory. In general, the agreement is significantly better for TNO/MACC than for EDGAR, except for the waste sector. The EDGAR

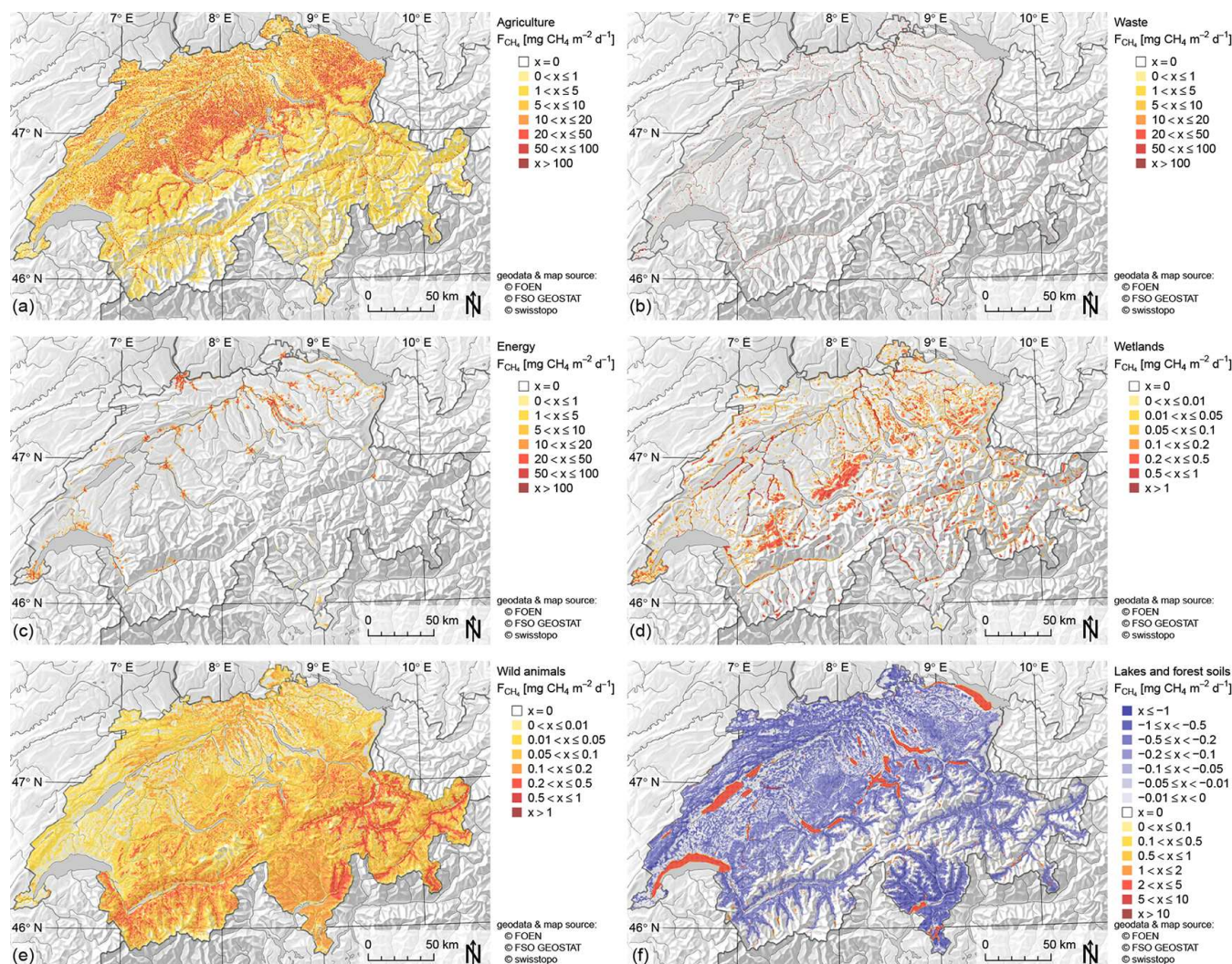


Fig. 2. Individual layers of the inventory presented in Fig. 1. Note that the scale for anthropogenic fluxes of the agricultural sector (a), the waste sector (b) and the energy sector (c) are a factor 10 to 100 larger than that for natural and semi-natural fluxes from wetlands (d), wild animals (e), and forest soil and lakes (f).

inventory tends towards higher fluxes as compared to our inventory, especially for the waste and energy sectors. EDGAR emissions for waste (30 Gg CH₄ yr⁻¹) and energy (44 Gg CH₄ yr⁻¹) are substantially higher than in the SGHG inventory (waste: 17 Gg CH₄ yr⁻¹, energy: 13 Gg CH₄ yr⁻¹). The relative difference for agricultural emissions is smaller, 162 Gg CH₄ yr⁻¹ for EDGAR compared with 153 Gg CH₄ yr⁻¹ in the SGHG. The TNO/MACC inventory compares well also at the sectorial level (agriculture: 163 Gg CH₄ yr⁻¹, waste: 17 Gg CH₄ yr⁻¹, and energy: 10 Gg CH₄ yr⁻¹ in 2009) against SGHG (agriculture: 151 Gg CH₄ yr⁻¹, waste: 16 Gg CH₄ yr⁻¹, and energy: 13 Gg CH₄ yr⁻¹ in 2009). Emissions in the EDGAR inventory are higher in densely populated regions (see Fig. S1 for a population density map) but lower in agriculturally dominated regions compared with our inventory (Fig. 3c),

suggesting that EDGAR allocates emissions too strongly to population density, consistent with the much higher values assigned to fuel distribution and waste disposal. Spatial differences are less pronounced between the TNO/MACC inventory and our inventory (Fig. 3d). In particular, the TNO/MACC inventory correctly identifies the regions of farming in the southern parts of the Swiss Plateau, but the emissions tend to be higher in these areas and lower in the mountains compared with our inventory. These spatial differences are further assessed in Sect. 3.3.1 to obtain a rough estimate of the uncertainty associated with the spatial disaggregation.

For natural CH₄ emissions, we only compare our country totals with numbers reported in an earlier study for Switzerland (SAEFL, 1996). Compared with that study, our estimates are considerably lower. Forests were considered

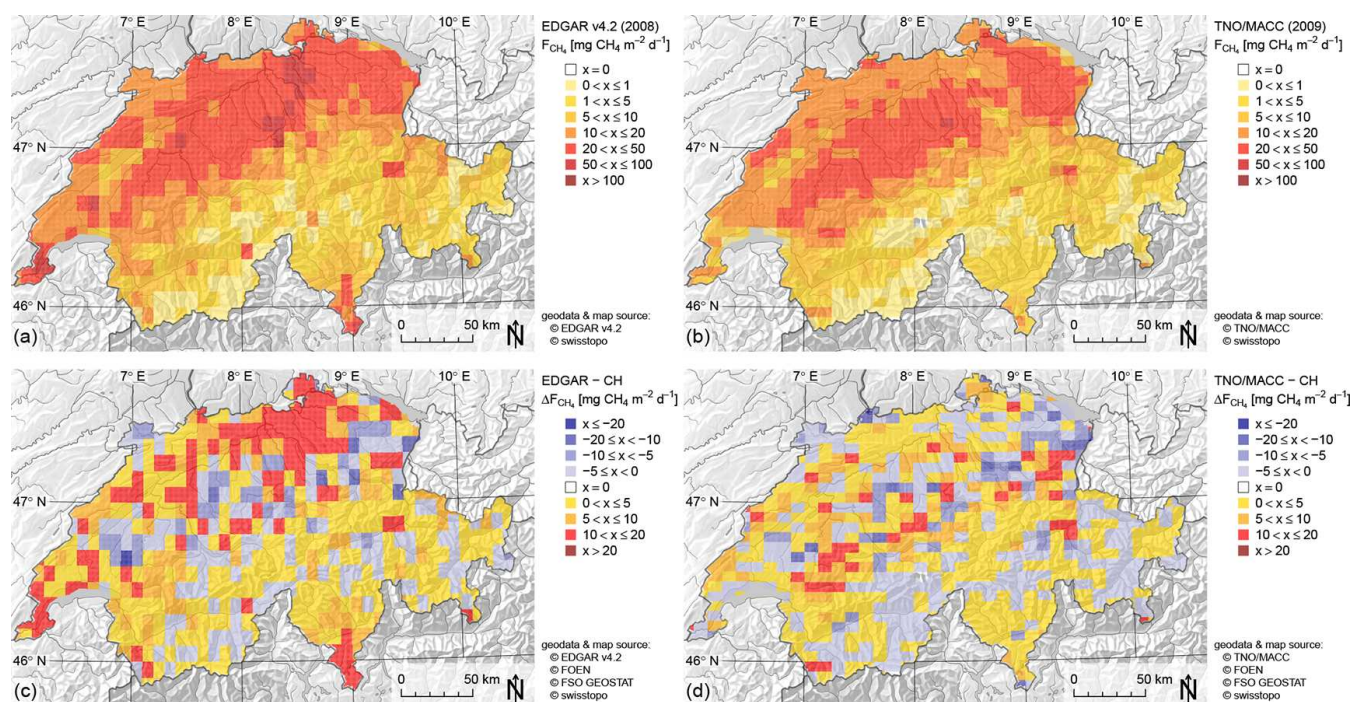


Fig. 3. Total anthropogenic CH₄ emissions over Switzerland according to the EDGAR v4.2 inventory for the year 2008 (a) and the TNO/MACC inventory for the year 2009 (b). Panels (c) and (d) are absolute differences from the total anthropogenic emissions in our inventory (Fig. 1).

a significant CH₄ source (50 Gg CH₄ yr⁻¹) in the former study, which was based on much more limited information. However, in the past two decades, no evidence for such strong CH₄ emissions could be found in Switzerland, and hence our updated estimate suggests that forests are a net CH₄ sink instead (net flux of -2.3 to -3.2 Gg CH₄ yr⁻¹) (Hobi, 2011). Moreover, contributions from small wild animals, namely rodents, are estimated to be much lower (0.027 Gg CH₄ yr⁻¹) than previously (2.8 Gg CH₄ yr⁻¹). Our findings indicate that agricultural soils may act as a small net sink (net flux of -1.5 to 0 Gg CH₄ yr⁻¹), while emissions of up to 2.1 Gg CH₄ yr⁻¹ were previously attributed to this type of ecosystem. In contrast, lakes had been estimated to be CH₄ neutral, whereas our findings suggest that they are a source of 2.3 Gg CH₄ yr⁻¹. Only the previous estimates for large wild animals (0.9 Gg CH₄ yr⁻¹) and wetlands (1.2 Gg CH₄ yr⁻¹) compare well with our study (1.1 Gg CH₄ yr⁻¹ and 2.3 Gg CH₄ yr⁻¹, respectively). Overall, the natural and semi-natural CH₄ emissions estimated in our study (5.7 Gg CH₄ yr⁻¹) are only about 10 % of those reported by SAEFL (1996), but equate to 3 % of the total CH₄ emissions in Switzerland.

3.3 Uncertainties of the inventory

For many purposes, and in particular for inverse modeling studies in which emission inventories are used as a priori es-

timates, it is important to quantify not only the distribution of the emissions but also their uncertainty. In the SGHGI (FOEN, 2013), an uncertainty is determined for each emission category based on errors associated with the activity data and the EFs. The combined uncertainties are listed in Table 2 together with the mean emissions for 2011. The uncertainty of the annual total emissions can then be computed as the square root of the sum of squares of the individual uncertainties, assuming uncorrelated errors. The uncertainty of the total Swiss anthropogenic CH₄ emissions is estimated to only 16 % (see Table 2), which is largely due to the low uncertainty of 18 % assigned by the SGHGI to the main emission source, 4.A Enteric Fermentation. It is interesting to note that this uncertainty is smaller than the difference between the SGHGI and the EDGAR v4.2 inventory.

For the uncertainty of emissions of a given grid cell at a given time, additional errors need to be considered, including errors associated with the spatial disaggregation and with the temporal variability as described in the following.

3.3.1 Spatial uncertainty

Uncertainties associated with the spatial disaggregation are difficult to assess. They depend on the accuracy of the spatial data sets, on quantization errors due to the use of discrete classes, on the relative weights assigned to individual sources, and on the often crude assumptions made for spatial

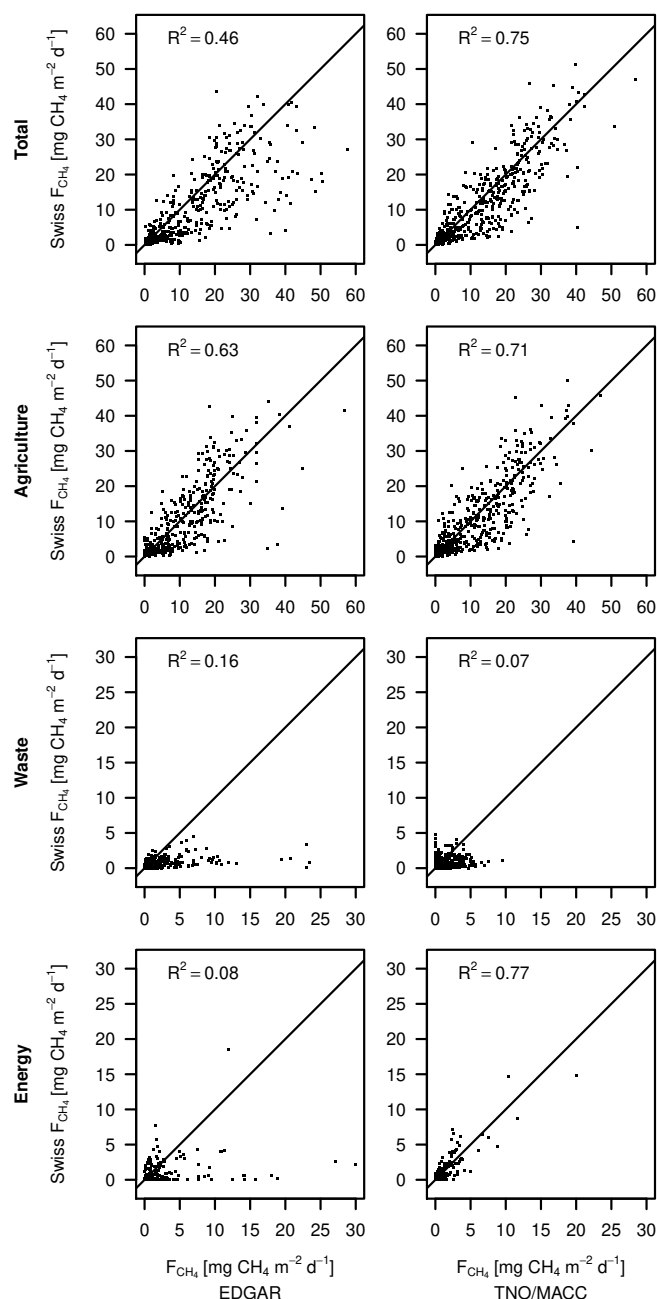


Fig. 4. Scatterplot between all pixel values in the EDGAR v4.2 and TNO/MACC inventories in Switzerland versus our inventory reduced to the respective resolution of the coarser inventory. Plots are shown for total emissions as well as for the sectors agricultural (EDGAR: IPCC_4A, IPCC4B and TNO/MACC: SNAP code 10), waste (EDGAR: IPCC_6A_6C, IPCC_6B and TNO/MACC: SNAP code 9) and energy (EDGAR: IPCC_1B2b and TNO/MACC: SNAP code 5) separately. The solid lines indicate the 1:1 relationship. The panels were scaled to show as much detail as possible and hence a few very large emissions were omitted.

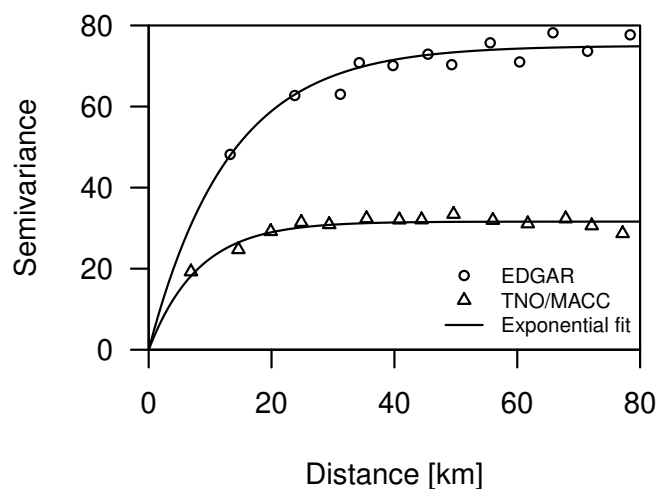


Fig. 5. Semivariogram of the differences between the EDGAR v4.2 and TNO/MACC inventories and our inventory. Also shown are exponential fits to the data (see text for further details).

disaggregation. Here we try to quantify the uncertainty of the emissions at the grid-cell level together with an error correlation length scale in a way that is consistent with the uncertainty of the country total of 16 %. For simplicity, uncertainties are only specified in terms of relative uncertainty of the grid cell total, but no distinction is made between different categories contributing to the total. The error covariance matrix C , which is an important input for inverse emission estimation, can then easily be formulated with diagonal elements

$$C_{ii} = (f E_i)^2 \quad (1)$$

and off-diagonal elements

$$C_{ij} = f E_i f E_j e^{-h/L} \quad (2)$$

where E_i is the total emission in grid cell i (the 2-D grid cell indices are combined here into a single index i), f is the relative uncertainty, h is the horizontal distance between grid cells i and j , and L is the error correlation length scale. The total emission of the country is given by

$$E_{\text{tot}} = \sum_i E_i \quad (3)$$

and the uncertainty of the total by

$$\sigma(E_{\text{tot}}) = \sqrt{\sum_i \sum_j C_{ij}}. \quad (4)$$

The error correlation length L was determined by comparing the spatial representation of the emissions in our inventory with that in EDGAR and TNO/MACC, all scaled to the same country total. The relative differences between the inventories are thus assumed to be a measure of the uncertainty

associated with the spatial disaggregation. The relative uncertainty factor f was finally chosen so that the relative uncertainty $\sigma(E_{\text{tot}})/E_{\text{tot}}$ is 16 %. To determine a representative error correlation length scale, we analyzed the variogram of the residuals R , where $R = E(\text{this study}) - E(\text{REF})$, the difference between emissions in our spatially explicit inventory $E(\text{this study})$ and emissions in a reference inventory $E(\text{REF})$ (EDGAR or TNO/MACC) (Fig. 5). A variogram describes the variance of the difference of a spatial variable R , i.e., $\text{var}(R(x) - R(x+h))$ as a function of the distance h (Cressie, 1993). The standard deviation of the residuals is larger for EDGAR than for TNO/MACC, which also results in a higher sill (see Fig. 5). As described in Lin and Gerbig (2005), a correlation length scale L can then be derived by fitting an exponential variogram model to the raw variogram. The length scale L obtained in this way was 13.0 km for EDGAR v4.2 and 8.0 km for TNO/MACC, which is close to the grid sizes of the two inventories, suggesting that their limited resolution is a constraining factor and that the true correlation length may be even smaller.

Assuming that the smaller correlation length scale of 8 km is more realistic, we obtain a value for the relative uncertainty f of 130 %. Emissions in individual grid cells thus have a large uncertainty and could well be double or half as large as estimated.

3.3.2 Temporal variability

Our spatially explicit inventory only includes annual mean emissions, but no seasonal and diurnal cycles due to a lack of suitable data. Nevertheless, we will briefly discuss the available temporal information relevant to our inventory to estimate the importance of temporal variability.

In the agricultural sector, livestock numbers are reported once a year in April, and seasonal fluctuations are only on the order of ± 3 %, with census data slightly above the annual mean (Bretscher, 2010). Within the traditional Swiss three-stage farming system, cattle are moved to Alpine meadows in summer to save the fertile valley floor for crop and winter fodder production. Hence, the spatial allocation of CH₄ emissions from ruminants changes between summer and winter. In addition, CH₄ emissions from ruminants depend on animal metabolism; thus, emissions peak following feed intake with a delay of a few hours and therefore display a diurnal pattern. Kinsman et al. (1995), for example, reported an average 20 % higher emission during the day than during the night, while Gao et al. (2011a, b) observed a diurnal peak-to-peak variation of up to a factor of two following the feeding rhythm. Seasonal CH₄ flux variations of about 20 % were attributed to lower emissions from manure at lower temperatures.

Emissions from manure are lowest at low temperatures and increase with longer storage duration, peaking only after about two months (Hindrichsen et al., 2005, 2006; Klevenhusen et al., 2010). The storage period before application is typically longer in winter, but lower storage temperatures likely dominate the influence on CH₄ production. Manure storage practice further influences CH₄ emissions (Külling et al., 2001, 2002, 2003). Higher than average emissions from farmyard manure are compensated for by lower emissions from urine-rich slurry, and on average do not differ significantly compared with complete slurry. Hence, these differences are of no relevance compared with the effects of storage period and temperature.

Landfills in Switzerland are covered by a soil layer; therefore, no large seasonal temperature fluctuations are expected within the deposited waste. However, the cover soil, where CH₄ is oxidized by methanotrophs, undergoes seasonal temperature fluctuations. More importantly, moisture positively influences CH₄ production in the waste body and inhibits CH₄ uptake in the cover soil (Chanton and Liptay, 2000). Both factors are expected to lead to higher CH₄ emissions in winter than in summer (Klusman and Dick, 2000). Nevertheless, the available information is insufficient to quantify seasonal fluctuations of CH₄ emissions from Swiss landfills in general.

A large proportion of natural gas is used for heating and therefore consumption is more than four times higher in January than in July (VSG, 2012). CH₄ emissions from leaking pipelines are therefore expected to be higher in winter than in summer, but no reliable data are available for proposing a seasonal cycle of these emissions.

Lake CH₄ emissions may exhibit a strong seasonal cycle similar to that found in a Swiss reservoir, where emissions were positively correlated with water temperature (DelSontro et al., 2010). In addition, turnover of a seasonally stratified water column can also contribute significantly to the annual CH₄ emissions from lakes (Schubert et al., 2010), a quite common process in Swiss lakes. Thus, both processes lead to a pronounced seasonal cycle in CH₄ emissions from water bodies in temperate zones, with higher emissions during summer than during winter (Sect. 2.2.1).

The above-listed diurnal and seasonal cycles indicate that observed CH₄ fluxes on a single day at a given time may differ significantly from the annual mean fluxes reported in our inventory, although the variations are probably smaller than those of other trace gases such as NO_x, which are dominated by traffic, heating, and other strongly varying activities. For the main emission sources (agriculture, landfills, gas losses) the available information is, unfortunately, not sufficient to provide specific time functions. In the absence of such information, inverse modeling studies need to make assumptions about the potential amplitude and correlation structure of such variations to specify realistic a priori uncertainties.

3.3.3 Scale dependency

When processing a spatially explicit inventory, a spatial resolution needs to be defined. For practical reasons it was decided to use a resolution of 500 m × 500 m, which is considerably better than the resolutions of a few kilometers typically used in regional-scale inverse modeling. Note that the input data were usually available at a higher resolution of 100 m × 100 m, and estimates were first obtained for this higher resolution and then averaged to the 500 m × 500 m grid. In many cases, the specific choice of spatial scale does not affect the total emissions estimated for a country. This is true for example of ruminant emissions where the total only depends on the number of ruminants but not on the granularity of their distribution within the country. Emissions from ecosystems such as wetlands, however, may critically depend on the chosen scale, in particular if they are estimated with an ecosystem model where the fluxes depend non-linearly on the spatial resolution of the input data. This was shown for example by Zhu et al. (2013), who found 42 % higher wetland CH₄ emissions when running their biogeochemistry model at 5 km × 5 km resolution as compared to a simulation at 100 km × 100 km resolution. They explained the difference by subgrid-scale variations in the water table that are smoothed out at a lower spatial resolution. In our case, we do not rely on a biogeochemistry model but on estimates of emission fluxes determined for specific wetland ecosystems, multiplied by the areas covered by these ecosystems in each grid cell. Nevertheless, these estimates may be biased if they are based on non-representative emission fluxes. Determining representative fluxes is particularly difficult for ecosystems, where the fluxes may vary on very small spatial scales. For example, DelSontro (2011) observed CH₄ flux variations across Lake Wohlen covering more than two orders of magnitude in CH₄ flux within a few tens of meters. It is therefore of great importance to combine individual flux measurements into values that are representative of the whole ecosystem (in this case a lake) before using them in an inventory. When finally building a gridded inventory, methods should be avoided that introduce artificial scale dependencies. Such problems are generated for instance when grid cells are assigned to a single land-use type, as shown, e.g., by Zhao and Liu (2013), but can be avoided using for example a mosaic approach that divides each cell into fractional contributions of all land-use classes (Mahrt and Sun (1995), Avissar and Pielke, 1989). We largely avoided such scale dependencies by first determining the land-use coverage of individual ecosystems based on high-resolution (100 m × 100 m or better) data sets before distributing the corresponding fluxes over the grid cells of the inventory.

3.3.4 Needs for building more realistic regional and national inventories

Regional and national inventories rely on spatial and temporal integration of local fluxes. Many studies are performed at a local scale to investigate the driving processes and often only cover short time periods of a few weeks or are only made in specific seasons. To obtain more representative results, we strongly recommend extending such measurements to complete years or even multiple years to include intra- and inter-annual variations. Even though fluxes deviating from the mean are more interesting for understanding the underlying processes, information on representative fluxes for given ecosystems is more valuable for building realistic inventories. The development of process-based models that result in spatially resolved flux estimates may also contribute to better inventories. At the same time, measurements integrating CH₄ fluxes over larger areas approximating the spatial resolution of the inventory are preferred for deriving emission factors. These recommendations are not limited to natural fluxes. Emission factors to estimate anthropogenic methane fluxes rely in the majority of cases on laboratory measurements. The total methane emissions of, e.g., a plant are rarely measured directly. Such direct measurements are especially desirable for revealing the true emission of sectors where different emission estimates result in large differences. Knowing the true total emissions would also help to investigate the efficiency of mitigation measures, as certain pathways might be missed when only investigating the known individual methane-producing processes.

4 Conclusions

A spatially explicit high-resolution CH₄ inventory was developed for Switzerland for the year 2011. This is the first comprehensive inventory at national level synthesizing most of the available Swiss data sets on anthropogenic as well as on natural and semi-natural fluxes. Anthropogenic emissions of 177 Gg CH₄ yr⁻¹ in 2011 are by far larger than the emissions from all natural and semi-natural sources, which were estimated to only 5.7 Gg CH₄ yr⁻¹, an order of magnitude less than an estimate reported in an earlier study on natural sources in Switzerland (SAEFL, 1996). Forest soils are estimated to be a net sink with a net flux of -2.8 Gg CH₄ yr⁻¹ and agricultural soils are estimated to be CH₄ neutral or a small sink with a net flux between -1.5 and 0 Gg CH₄ yr⁻¹ for agricultural soils, partially offsetting the natural emissions. In total, Switzerland acted as a net CH₄ source of 180 Gg CH₄ yr⁻¹ in 2011. With a share of nearly 85 %, agricultural emissions are by far the most important anthropogenic source in Switzerland, followed by the waste and energy sectors. The uncertainty of the total anthropogenic emissions is estimated to be only 16 %, which is largely a result of the low uncertainty assigned to the largest single CH₄ source

– enteric fermentation of ruminants. Detailed geospatial information is available for Switzerland, thereby allowing the spatial allocation of the individual emission sources. Information on temporal variability of CH₄ emissions, however, is very sparse and currently insufficient for prescribing diurnal and seasonal variations, an aspect that should be better addressed in future studies.

This inventory will provide invaluable input for regional-scale atmospheric modeling and inverse source estimation, which are urgently needed for independent validation of inventories based on atmospheric measurements. The spatial disaggregation of other CH₄ sources currently not covered by this inventory, especially from biogas production and composting, might become more critical in the future with the expected increase in the relative importance of these sources. The methodology suggested here is not specific to Switzerland and could be used to derive similar inventories for other countries, provided that country-specific emission factors are used where necessary and detailed geostatistical data are available.

Supplementary material related to this article is available online at <http://www.biogeosciences.net/11/1941/2014/bg-11-1941-2014-supplement.pdf>.

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